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SAVANNAH RIVER LABORATORY

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M E M O R A N D U M

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FABRICATION AND MECHANICAL TESTING OF Pd-Sm_2O_3 CERMET WIRE

Summary

This report provides estimates of the mechanical properties of Pd-^{252}Cf oxide cermet wire as indicated by measurements on Pd-Sm_2O_3 cermet. In the course of fabricating wires to measure the tensile properties of Pd-Sm_2O_3 cermet, fabrication techniques were developed to double the previously allowable oxide content to at least 2.5 v/o total oxide for a 0.040-inch-square rolled wire. This loading is equivalent to 1 mg ^{252}Cf plus five-fold additional feed impurities per inch of 0.040-inch-square wire; this is twice the proposed maximum specification for bare wire and provides lattitude to increase the ^{252}Cf concentration in cermet cores to meet the same linear concentration in clad wire of the same diameter. Splintering sometimes occurs during fabrication of wire with >2.5 v/o oxide. The improved fabricability is achieved by increasing the density of the pellet from about 60% to 85% by pressing at 14,000 psi, heating up to sintering temperature over two hours, and sintering for two hours at 1300°C.

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Fabricated cermet wires have physical properties typical of powder metallurgy products of noble metals. Ductility ranges between 3% and 14%. The ultimate tensile strength increases from 23,000 to 41,000 psi with an increase in oxide concentration to 5 volume percent oxide. Yield strength ranges between 9,000 and 17,000 psi and is not affected by oxide concentration. The hardness of cermet containing >1.25 v/o oxide is 80-90 DPH and is not affected by oxide concentration.

Introduction

Wire containing up to 0.5 mg ^{252}Cf per inch is being developed as an alternative product to Cf_2O_3 or $(CfO)_2SO_4$. Such wire can be subdivided by the encapsulator to make several sources of the desired size, thus avoiding the wet chemistry involved in handling oxide or oxysulfate. The wire is made as a cermet in which the particles of Cf_2O_3 are dispersed in a matrix of pure Pd. The oxide phase used in development work was Sm_2O_3. In the production of wire for sale, the oxide phase will be a mixed oxide of ^{252}Cf, terbium carrier, and impurities associated with ^{252}Cf, containing typically 20% ^{252}Cf (SRO-153). A chemical plating process has been developed for producing the Pd-^{252}Cf oxide cermet which can be fabricated into wires containing uniform linear concentrations of 5, 50, and 500 μg ^{252}Cf per inch of 0.040-inch-square wire.

Initially, cermet wires containing over 1 v/o oxide, or .4 mg ^{252}Cf per inch of 0.040-inch-square wire, could not regularly be formed by rolling from pellets; many wires cracked and fractured during the early rolling steps because of low ductility until the Pd matrix was continuously bonded. Reactor Engineering Division is developing processes to produce wire on a larger scale and to apply cladding by swaging and drawing techniques. Early efforts to swage-clad cermet wire showed severe breakup of the cermet core presumably, in view of this work, caused primarily by low density of the cermet billets.

Consequently, a study of the fabrication variables and mechanical properties was undertaken to increase the oxide content that could be accommodated by rolling cermet pellets made the chemical plating process, to assist RED in matching properties of clad and core materials to develop swaging and drawing techniques for clad wire, to provide a base line for determining He aging effects on the cermet wire, and to provide information on the mechanical properties as a guide to encapsulators in establishing their processes.

Basic Fabrication Process

The basic process for fabricating Pd-^{252}Cf oxide cermet wire is to admix by chemical plating Pd onto oxide, form a pellet by powder metallurgy techniques, and roll into wire. The ^{252}Cf feed is leached from the shipping package into a plating flask in the form of a nitrate solution. Californium oxalate is precipitated with oxalic acid and digested. Hydrazine reductant and a palladium salt solution are added to the suspended precipitate. The hydrazine reduces the Pd ions to metal which coats the Cf oxalate particles. The coated particles are filtered, rinsed, dried, and pulverized. The powder is calcined at 450°C in a 4% H_2-96% He atmosphere for thirty minutes, cooled, and pressed. The resulting pellet is sintered at 1300°C in argon and alternately rolled and annealed to form about 3-3/8 inches of 0.040-inch-square wire per gram of Pd.

Effect of Powder Metallurgy Process Variables on Fabricability of Wire

The porosity of the sintered pellet, rather than dispersion hardening, by the oxide particles, appears to be the factor that limited the fabricability of cermet containing less than 2.5 v/o oxide. In addition, density gradients (laminations) formed by compacting at too high a pressure caused transverse cracking during rolling. Pellets sintered for two hours rather than thirty minutes showed an improvement in density and rollability. Pellets pressed at 14,000 psi instead of 40,000 psi showed a decrease in the number of failures by transverse cracking. These observations are supported by the data in Table I.

Eighteen pellets of varying oxide concentration were pressed at 40,000 psi and sintered in an argon atmosphere for thirty minutes at 1300°C with a thirty to forty-five minute heatup to sintering temperature (Table I). Only six of these pellets could be rolled into acceptable wires. Seven failed by transverse fracture and five by longitudinal fracture. Six of those which fractured transversely contained $\leqslant$1.25 v/o oxide. All of the longitudinally fractured pellets contained at least 2.5 v/o oxide.

By lowering the pressing load from 40,000 psi to 14,000 psi and by lengthening the heating time to sintering temperature to two hours, the number of failures was greatly reduced. Of nine pellets, only three failed by transverse cracking, none by longitudinal cracking, and one by splintering. (A wire was formed but it splintered during the last pass through the rolling mill.) The density range of sintered pellets containing 1.25 v/o oxide increased from 53-63% to 57-73%. Densities for sintered compacts containing 1.87 and 2.50 v/o oxide ranged from 78-87% and from 86-88% of theoretical, respectively.

Densities increased and the number of failures decreased when, in addition to pressing at 14,000 psi and taking 2 hours to reach the sintering temperature, pellets were sintered 2 hours at 1300°C rather than 30 minutes (Table I). The maximum density obtained was 89% of theoretical for a single pellet containing 2.5 v/o oxide; the minimum was 75% for a pellet containing 5.0 v/o oxide. One pellet containing 1.87 v/o oxide failed during rolling because it contained quartz fragments from a devitrified annealing tube. Four pellets containing 5.0 v/o oxide splintered but all were rolled to 0.040-inch-square wire.

Types of Failures During Rolling

Transverse cracking, longitudinal cracking, and splintering were the types of failures observed during this study. Transverse cracking (Figure 1a) occurred in pellets that were pressed at a high load, causing lamination. These laminations created a low density area in the pellet which did not heal with thirty-minute sintering. A fast heatup to sintering temperature closed the pores of the pellet and trapped gases in the pellets when pressed at high loads. Thus, a longer time was required for the gas to diffuse from the compact.

Longitudinal cracking (Figure 1b) also occurred when a low density area was created. In addition, longitudinal fracture occurred when the cross sectional area of a low density pellet was reduced by more than 15%, especially when the cross section of the pellet was rolled from round to square with the associated large radial translations. Oxide content had an effect on longitudinal fracture. Only those pellets containing at least 2.5 v/o oxide failed longitudinally. Except for the wire at 1.87% v/o oxide that contained quartz fragments, no longitudinal fractures occurred at <2.5 v/o oxide.

Splintering occurred in those wires containing at least 5 v/o oxide. The wires could be rolled to 0.040 inch, but appeared very much like the 12.5 v/o oxide wire shown in Figure 1c.

Mechanical Properties

All fabricated wires that were not splintered were tested after annealing for 10 minutes at 850°C on an Instron tensile test machine to measure the ductility (Figure 4), yield strength (Figure 5), and ultimate strength (Figure 6) of the cermet. Most of the data is for a gage length of ¼ inch. The number of tests at each oxide content is indicated in the circle and the range of the data is shown by the vertical bars. Data for wires that failed within the grips were discarded. All wires failed in a ductile manner, as shown in Figures 2 and 3; Figure 2 is a photograph of a Pd-1.87 v/o Sm_2O_3 wire showing a necked-down region near the fractured end typical of ductile failure. The fractures were "cup-and-cone", an indication of ductile failure. Figure 3 shows the fractured surface of a Pd-1.25 v/o Sm_2O_3 wire.

The ductility of the cermet wires ranged between 3 and 14% (Figure 4). No direct correlation could be distinguished between ductility and oxide content. Bar stock Pd which had been swaged to 0.040 inch diameter had a ductility of 5%. An elongation of 5% was also observed for the powder metallurgy product of the plating process which contained no oxide. Both of these forms thus exhibited a lower ductility than the oxide-loaded Pd.

The yield strength, which is the stress at the beginning of plastic deformation, ranged from 9,000 and 17,000 psi for cermet wire and was not affected by oxide content (Figure 5).

The ultimate tensile strength, which when compared to the yield strength is a measure of the work hardening of a metal, appeared to increase from 20,000 psi to 40,000 psi with an increase in oxide content to 5 v/o (Figure 6). The range of tensile strength for Pd wire swaged from Pd rod was 29,000 to 39,000 psi. The tensile strength for wire made by chemical plating with no oxide was 23,000 psi.

The hardness of wires containing $\geqslant$1.25 v/o oxide ranged between 80 and 90 DPH compared to about 50 for pure Pd and was not affected by oxide content above 1.25 v/o oxide (Figure 7).

Table II lists values for the ductility, tensile strength, and yield strength of dispersion hardened gold and platinum.[1] In contrast to Sm_2O_3-Pd cermets, the ductility of Pt-ThO_2 cermet is lower than for pure Pt and increases with an increase in oxide content; the tensile strength is higher than for pure Pt, but is not significantly affected by oxide content. In general, the ductility, tensile strength and yield strength for Au-Al_2O_3, Pt-ThO_2 and Pd-Sm_2O_3 are about the same, indicating that the Pd-^{252}Cf oxide cermet wires should have mechanical properties typical of these powder metallurgy products.

Applicability of Observations on Pd-Sm_2O_3 Standin to Pd-^{252}Cf Oxide Cermet

Impurities in Cf feed equivalent to five times the Cf content can produce an oxide volume up to ten times the volume of Cf_2O_3. The fabricability limit for rolling is 2.5 v/o total oxide or .25 v/o Cf oxide, which is equivalent to 1 mg ^{252}Cf/inch of 0.040-inch-square wire. This concentration is twice the proposed maximum specification for bare wire and affords the even higher core concentrations required to achieve the maximum proposed specification of 500 µg ^{252}Cf/inch with clad wire.

The properties of Pd-Sm_2O_3 cermet wire should be typical of those for Pd-^{252}Cf oxide cermet wire except for radiation effects, specifically, fission damage and helium buildup from alpha decay. Alpha radiation effects can be simulated with ^{242}Cm which α-decays six times faster than ^{252}Cf. Tests are in progress to evaluate the effects of radiation embrittlement on cermet fabricability by rolling ^{242}Cm cermet or ^{252}Cf cermet wires to .050-inch - .100-inch cross section, storing these wires for several months, and then rolling them further to .030-inch - .040-inch cross section.

WCM:rbw

REFERENCE

1. N. Fuschillo and M. L. Gimpl, "Dispersion Hardened Platinum and Gold Alloys for Electrical Applications," Journal of Metals, June 1971, pp 39-40.

TABLE I

EFFECT OF $Pd-Sm_2O_3$ CONSOLIDATION PARAMETERS ON WIRE FABRICABILITY

Press: 40,000 psi

Sintering: 30-45 min. heat up
30 min. sinter @ 1300°C

v/o Oxide	Number Pellets	Number Wires	Failures Trans. Crack	Failures Long. Crack	Failures Splinter	Density(%)
.62	3	0	3			
1.25	6	3	3			53-63%
2.50	3	2		1		
5.00	6	1	1	4		

Press: 14,000 psi

Sintering: 2 hr. heat up and
30 min. sinter @ 1300°C

v/o Oxide	Number Pellets	Number Wires	Failures Trans. Crack	Failures Long. Crack	Failures Splinter	Density(%)
1.25	3	2	1		1	57-73%
1.87	3	1	2			78-87%
2.50	3	3				86-88%

Press: 14,000 psi

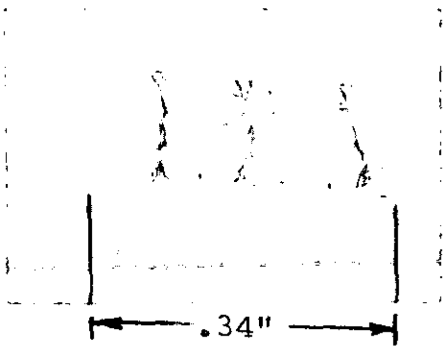
Sintering: 2 hr. heat up and
2 hrs. sinter @ 1300°C

v/o Oxide	Number Pellets	Number Wires	Failures Trans. Crack	Failures Long. Crack	Failures Splinter	Density(%)
1.87	3	2		1		80-86%
2.50	1	1				89%
5.00	3	2			1	75-85%
7.50	3	0			3	85-86%

TABLE II

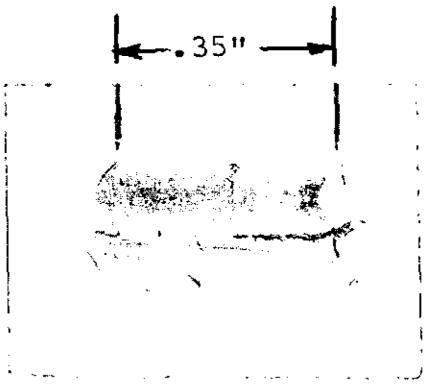
PHYSICAL PROPERTIES OF DISPERSION HARDENED GOLD AND PLATINUM[1]

Volume Percent Oxide	YS(psi)	UTS(psi)	Ductility(%)
Pt(cast)	--	18,000	40.0
Pt-1 v/o ThO_2	--	36,000	6.0
Pt-1.8 v/o ThO_2	--	39,500	12.0
Pt-2.2 v/o ThO_2	14,000	30,000	20.0
Au(cast)	--	--	25.0
Au-3.5 v/o Al_2O_3	10,000	16,500	12.0
Au-6.4 v/o Al_2O_3	9,100	17,200	11.5



.140" square Pd-1.87 v/o Sm_2O_3 pellet

a. Transverse Fracture



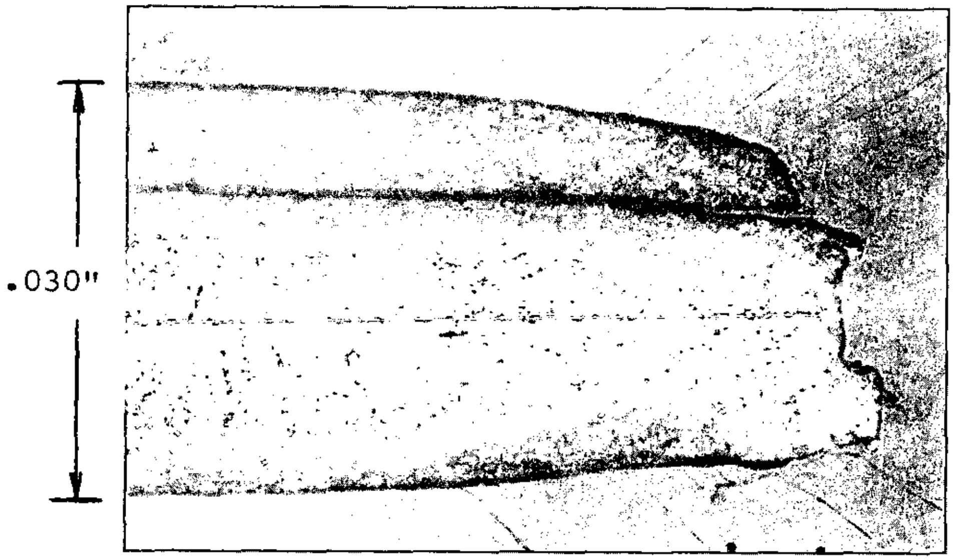
.125" square Pd-5 v/o Sm_2O_3 pellet

b. Longitudinal Fracture

.030" square Pd-12.5 v/o Sm_2O_3 wire

c. Splintering

FIG. 1: FAILURE TYPES FOR ROLLED Pd-Sm_2O_3 CERMET



Pd-1.87 v/o Sm_2O_3 Wire
7.9% elongation
Y.S. = 12,300 psi UTS = 34,000 psi

FIGURE 2: DUCTILE FAILURE MODE FOR Pd-Sm_2O_3 CERMET

.001"

Pd-1.25 v/o Sm_2O_3 Wire
4.6% elongation
Y.S. = 12,400 psi UTS - 26,900 psi

FIGURE 3: SCANNING ELECTRON MICROGRAPH OF A TYPICAL DUCTILE FRACTURE SURFACE FOR Pd-Sm_2O_3 CERMET

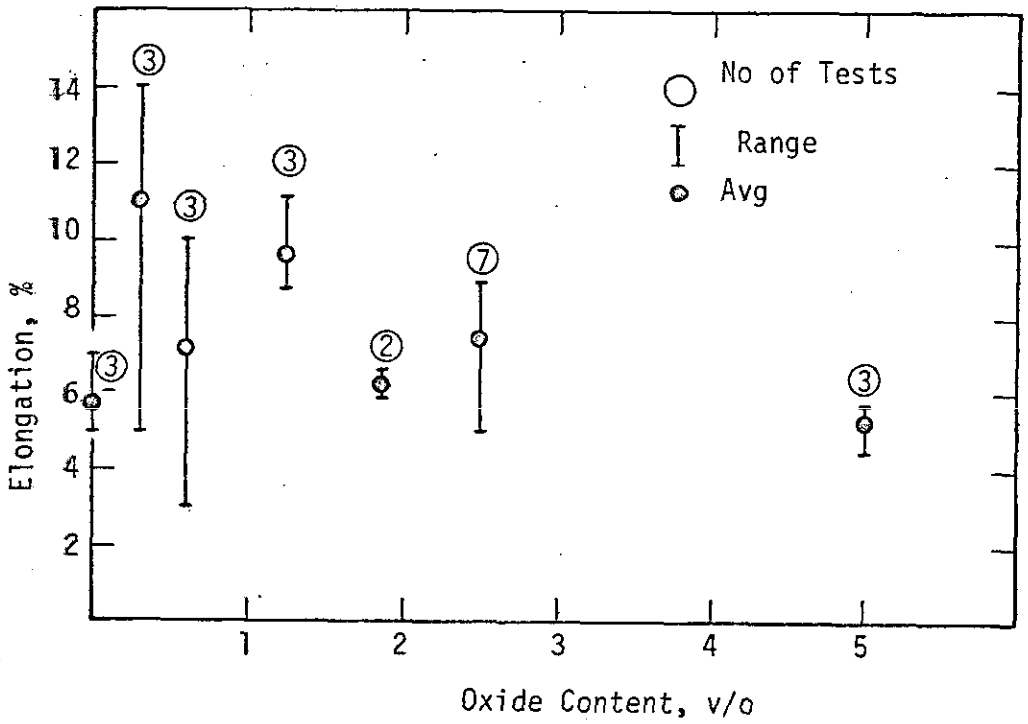


FIG. 4 Ductility of Pd-Sm_2O_3 Cermet

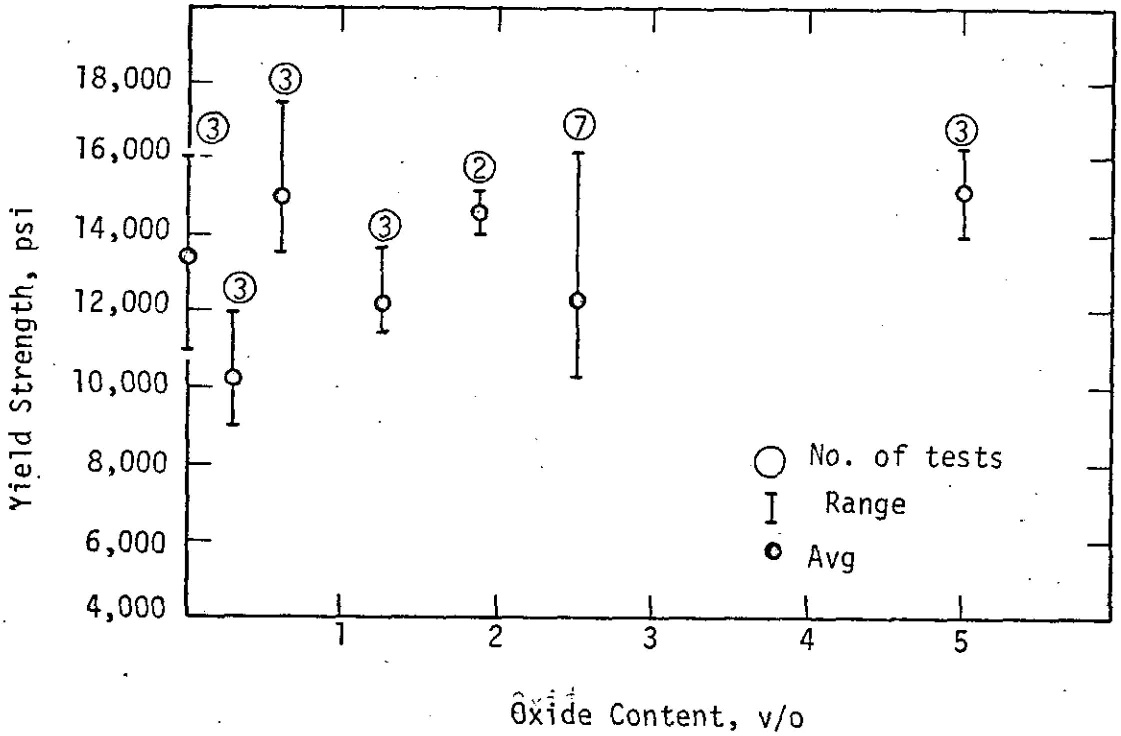


FIG. 5 Yield Strength of Pd-Sm_2O_3 Cermet

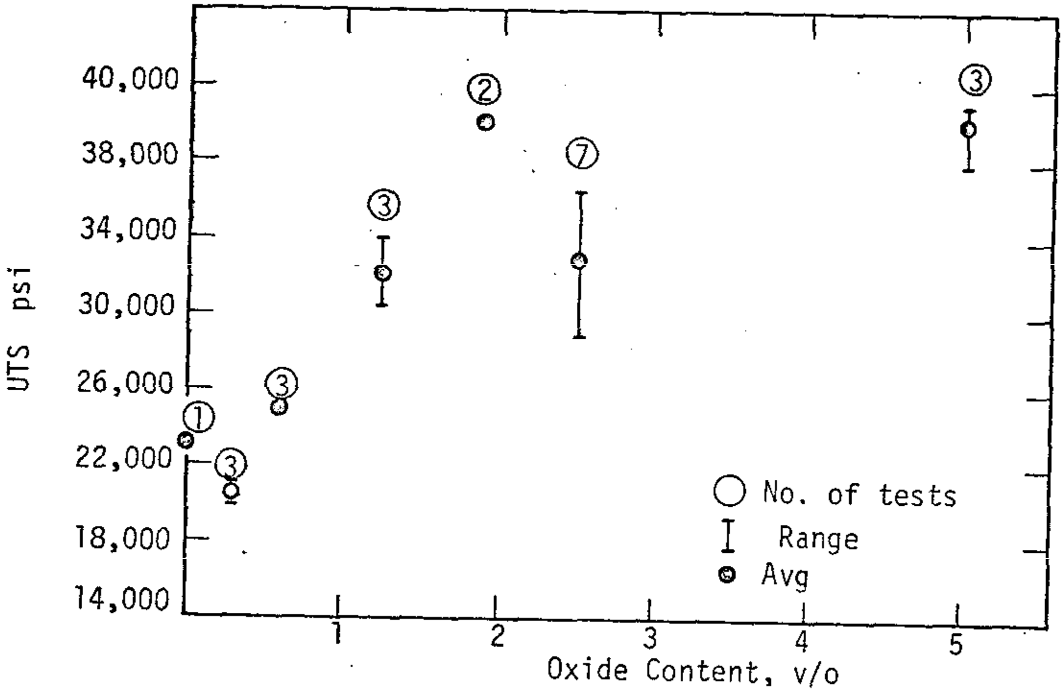


FIG. 6 Ultimate Strength of Pd-Sm_2O_3 Cermet

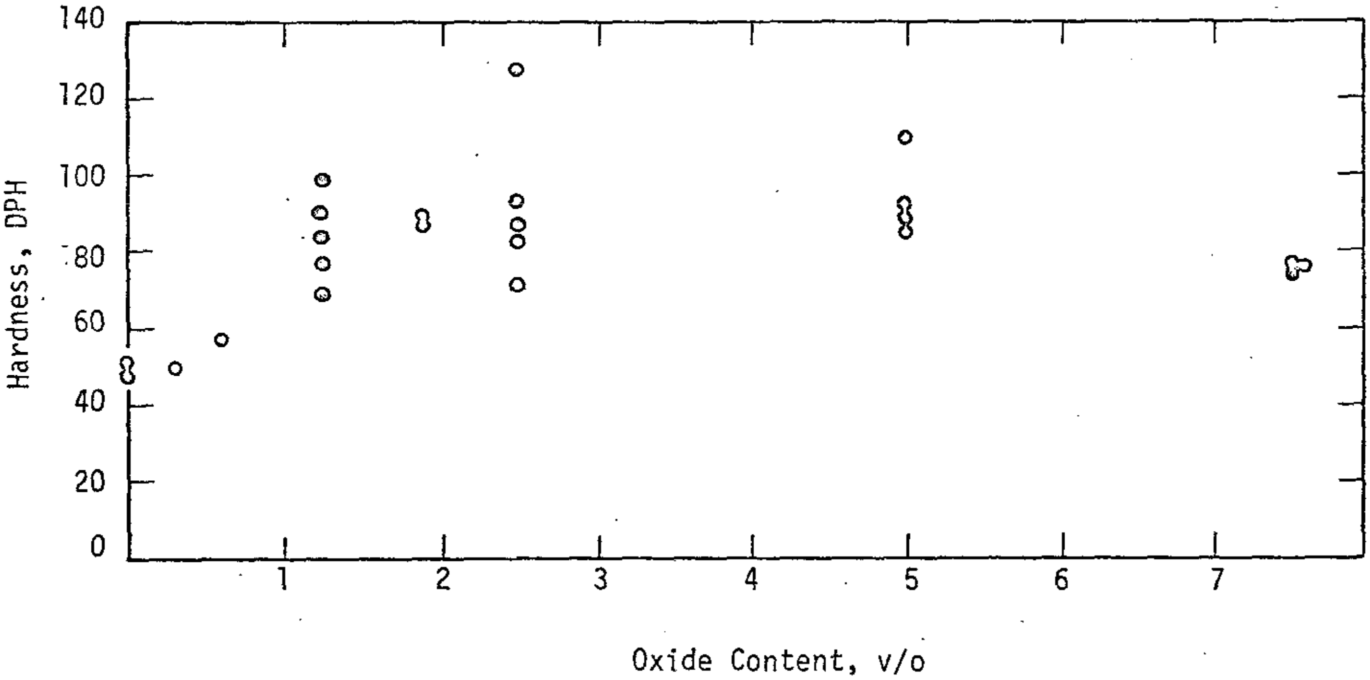


FIG. 7 Hardness of Pd-Sm_2O_3 Cermet